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2010 J. Phys.: Conf. Ser. 210 012041

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Surface photovoltage and photoluminescence spectroscopy of self-assembled InAs/InP quantum wires

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Abstract. The optical properties of InAs/InP multi-layer quantum wire (QWR) structures of various spacer thicknesses have been investigated by means of room temperature surface photovoltage and photoluminescence spectroscopy. Combined with empirical tight binding calculations, the spectra have revealed transitions assigned to QWR families with heights equal to integer number of 5, 6 and 7 monolayers. From the comparison of the experimental and theoretical results the atomic concentration of phosphorus in the wires has been estimated.

1. Introduction

Multi-layer structures with self-assembled InAs/InP quantum wires (QWRs) are promising candidates for light emitting devices in the 1.3 - 1.55 μm wavelength range used in fibre optical telecommunications [1]. The optical properties of InAs/InP QWRs have been studied mainly by low-temperature photoluminescence (PL) spectroscopy revealing several wide bands, which have been generally attributed to emission of QWRs belonging to different families having heights differing by an integer number of monolayers (MLs) [2-4]. Calculations of the electronic structure have been performed using the envelope function approximation (EFA) [2, 5]. Experimental investigations at room temperature (RT), which will be the operating temperature of a final device, are relatively few, including Raman [6] and optical absorbance spectroscopy [7]. An alternative to avoid the difficulties related to optical absorbance measurements in nanostructures is the surface photovoltage (SPV) spectroscopy, because of its high sensitivity even at RT [8-10]. In this work we apply the SPV spectroscopy to study multi-layer InAs/InP QWRs at RT in combination with PL measurements,

structural data from previous studies [4], and comparison with empirical tight binding (ETB) electronic structure calculations.

2. Experimental and calculation details

Four samples were grown on (001) semi-insulating InP:Fe substrates by solid-source molecular beam epitaxy at 515°C [4]. They contain a 150 nm InP buffer layer and a stack of seven InAs QWR layers, separated by InP spacers of thickness $d = 3, 5, 10$ and 20 nm in the four samples. The inset of figure 1(a) shows a schematic of the sample structure. The residual doping in the buffer and the structure is n-type, of the order of $1 \times 10^{17} \text{ cm}^{-3}$. Atomic force microscopy (AFM) [4] has shown that the QWRs are oriented along [1-10] and their average height H increases from 1.5 to 2.2 nm when d decreases from 20 to 3 nm. Both values are much smaller than the lateral QWR width, which is of the order of 13 nm.

The set-up used for SPV spectral measurements in metal-insulator-semiconductor operation mode is described elsewhere [10]. Here the semitransparent probe electrode was fixed at a small distance above the sample, which was glued by conductive silver paste to a grounded copper platform. The incident photon flux density was kept constant ($\Phi \approx 1.5 \times 10^{14} \text{ cm}^{-2} \text{ s}^{-1} \pm 0.5\%$) for each wavelength. All measurements were performed with normal incident light chopped at 94Hz, scanning from high toward low wavelengths. The PL was excited by the 514.5 nm line of an Ar⁺ ion laser, dispersed by a 0.19 TRIAX single monochromator (600 grooves/mm grating) and synchronously detected by a N₂-cooled InGaAs detector.

The procedure for calculating the electronic structure by means of the ETB method and the materials parameters are described in detail in [11]. The intermixing that inevitably exists in such structures was taken into account by considering a nonzero P concentration in the wire; it has been shown [11] that this method is equivalent to introducing a graded concentration profile. Since InAs/InP wires and dots are relatively flat, the effect of the lateral confinement on the transition energies is small [11] and was therefore neglected in our calculations. A temperature correction of 65 meV is subtracted from the calculated energies to account for the InAs band gap decrease between 0 and 300 K [12].

3. Results and discussion

In the range 0.75–1.1 eV the SPV amplitude increases with increasing the photon energy $h\nu$ revealing a few steps superimposed on a smooth background (see figure 1(a)). The origin of the latter is unknown: it could be related to deep level absorption or an Urbach tail below the InP bandgap. Though the Urbach tail is usually assumed to have exponential form, in our case the background is much better represented by a parabolic term similarly to [13]. Subtracting it from the SPV spectrum results in three SPV peaks at around 0.91, 0.96 and 1.01 eV as shown in figure 1(b). For the sample with $d = 3$ nm there are only two peaks at 0.93 and 0.99 eV. The SPV phase (not shown) is in the 4th quadrant, which indicates an upward energy band bending in the surface space charge region (SCR) with respect to the bulk [9]. This is in accordance with the n-type residual doping of the structure and buffer layer.

The RT PL spectra (figure 1(c)) reveal a broad band around 0.8 eV and another one at higher $h\nu$. For the samples with $d = 10$ and 20 nm the latter band can be decomposed into three Gaussians centred at about 0.91, 0.94, and 0.98 eV. Notice that the dip at 0.89 eV is an artifact of the gratings that does not allow precise positioning of the first Gaussian – it could be at a lower energy. However, the peaks can be traced back to lower temperatures, where they appear more clearly in the PL spectra. The energy positions of these PL features are in good agreement with the optical transitions observed by SPV. Since these features are absent in the PL spectrum of bulk InP (not shown) we can ascribe them to electronic transitions arising from the QWRs. The difference between the SPV and PL peak positions represents the Stokes shift for the QWR exciton. The obtained values (20-30 meV) agree with the reported one (26 meV) for InAs/InP QWRs [7].

The SPV signal from the QWRs is due to photocarriers, which thermally escape from the InAs potential wells into the InP barriers, where they are separated by the built-in electric field of the SCR. Since the carrier escape is easier for higher energy states, the SPV peaks at higher $h\nu$ are stronger.

Thus, the SPV spectra complement the information obtained by PL, where the lowest energy transition is predominantly observed.

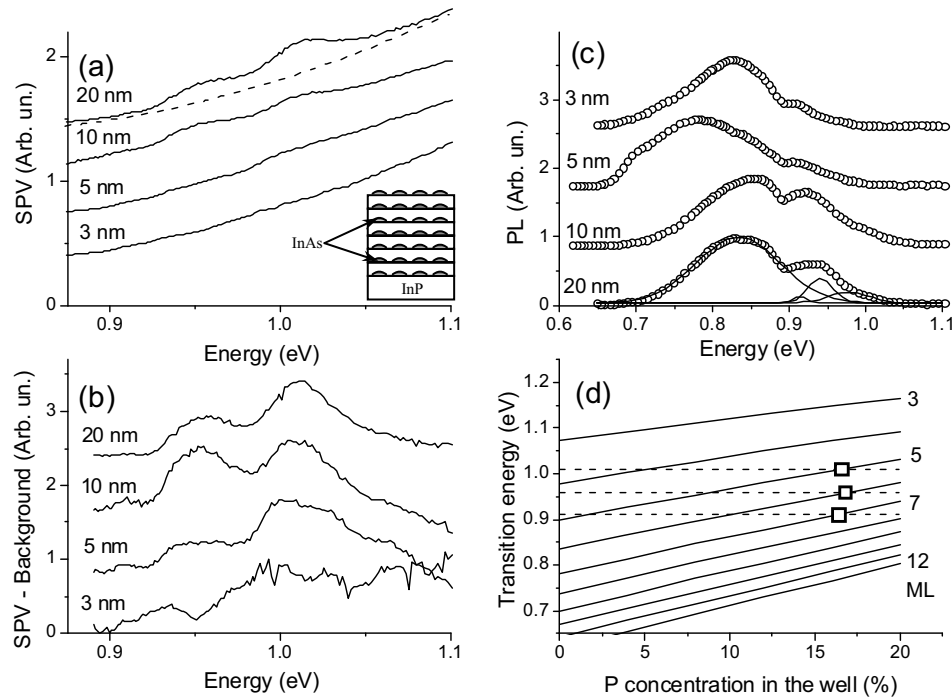


Figure 1. SPV (a and b) and PL (c) spectra of samples with spacer thickness 3, 5, 10, 20 nm. All curves are shifted vertically for clarity. The dashed line in (a) illustrates the background (for this sample), which is subtracted in (b). (d) Transition energies of a single InAsP/InP QWR as a function of the P content for wire heights from 3 to 12 MLs. The dashed lines show the SPV peak values and their relevant intersections with the calculated curves are illustrated by squares.

Following the discussion in previous works on InAs/InP QWRs [2-4] we ascribe the observed PL and SPV spectral bands to excitonic transitions that take place in different QWR families having heights differing by an integer number of MLs.

Figure 1(d) displays the calculated transition energies as a function of the P concentration [P] in a single QWR for different values of the wire height H , and the obtained dependences are linear. The experimental peak energies obtained by SPV are also shown with horizontal dashed lines. From the intersections between the two sets of lines the QWR heights and the P concentration are determined keeping in mind that i) H should increase by a step of 1 ML and ii) [P] should remain nearly constant for the different QWR families [11]. Thus the peaks at 0.91, 0.96 and 1.01 eV are identified as originating from excitonic transitions in QWR families with $H = 7, 6$ and 5 ML, respectively and with P concentration around 16%. These values of H agree with those obtained in [4] by low-temperature PL combined with EFA calculations [2]. The obtained value of [P] is in accordance with those found for the interface region of the same QWR samples ($\sim 30\%$) by Raman spectroscopy [6]. The error in [P] can be estimated to $\pm 4\%$ and results form the uncertainty in the InAs/InP valence band offset (0.3-0.4 eV, $\pm(2-3)\%$ [11]), the experimental error in the transition energies (± 5 meV, $\pm 0.6\%$) and the effect of the strain relaxation during the QWR formation ($< 1\%$ [11]).

The lowest energy PL peak at ~ 0.8 eV is much broader and could account for a number of families (8-12 ML). Alternatively, this emission could originate from larger QWRs formed by the overlapping of two or more small wires, as observed by transmission electron microscopy [14] and AFM [4].

With decreasing d the number of thicker QWRs increases as explained in [15]. Besides the electron coupling progressively widens the bound state energies into superlattice minibands, thus broadening the absorption (SPV) spectrum. As a result the large PL peak at low $h\nu$ increases at the expense of the high-energy ones, while the SPV steps become less pronounced. Also a slight red shift (~ 20 meV) of the SPV and PL spectral features is observed in figure 1 (b) and c) when d decreases from 20 to 3 nm. This shift is smaller than the one obtained by low-temperature PL (25-50 meV) in [4], which could be related to thermal population of nearby levels in the minibands. Due to the broad spectral shapes and the uncertainties in the peak positions in the RT spectra it is difficult to analyze this problem in detail.

It should be mentioned that the transitions of the $d=3$ nm sample, observed by SPV, are consistent with a lower P concentration (around 13%) than that of the other samples. This is in agreement with a decrease in the P content for thinner samples observed previously by Raman spectroscopy [6].

4. Conclusion

The optical absorption and emission of multi-layer InAs/InP QWRs at RT have been studied by SPV and PL spectroscopy, combined with structural data and ETB calculations. The existence of several QWR families with heights of 5, 6 and 7 MLs has been revealed, which supports previous findings by low-temperature PL. Further, the degree of P intermixing in the wires has been estimated to be equivalent to a constant P concentration of the order of $(16\pm 4)\%$ and an upward direction of the energy band bending at the surface is found in accordance with the residual doping in the structure. The study highlights the potential of the SPV spectroscopy for characterization of complex nanostructures at RT.

Acknowledgements.

This work was supported by the Sofia University research fund (contract 99/2009), the Bulgarian National Science Fund (contract D01-463/12.07.06), the Alexander von Humboldt Foundation, the Ministry of Education and Science of Spain (Project No. MAT2006-01825, FEDER), and the Generalitat Valenciana.

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